Analysis Tools and Techniques Program

The continuing shrinking of device dimensions and the rapid inclusion of new materials into device fabrication demands the development of new analytical tools and techniques. The need for new analytical techniques and tools has increased as the components in the transistors approach the low nanometer level and the number of transistors per chip approaches 1 billion. The development of tools capable of characterizing the structures produced in the laboratories as well as those needed to confirm the manufacturing process require significant improvements; although those used for production also require significant speeds not to slow down the commercial manufacturing. This latter condition may require some sacrifices in the resolution and accuracy of those tools. In addition, more significant modeling that allows us to bridge those areas where measurements can be done to those where knowledge is needed from, but measurements cannot be done are critical.

Thin-Film X-Ray Metrology For Microelectronics

Technical Contact:
James P. Cline
Donald Windover

Nicholas Armstrong
University of
Technology Sydney
Australia

Goals

This multi-year collaborative effort between SEMATECH and NIST will provide the semiconductor community with the methodology for accurate thin film characterization using X-Ray Reflectometry (XRR).

Customer Needs

In recent years, the semiconductor industry has driven scientific advancement towards processing nanometer-scale material coatings with unprecedented uniformity in thickness, composition control, and unique electrical and mechanical properties. Nanotechnology represents the fastest growth area of industry in the United States today. Simultaneous to this rapid advance in thin film processing technologies, the X-ray diffraction user community and instrument manufacturers have collaboratively developed techniques such as High-Resolution X-Ray Diffraction (HRXRD) and X-Ray Reflectometry (XRR) that permit the quantitative profiling of thin film characteristics, such as thickness, density profile, composition, roughness, and strain fields. With the XRR method in particular, parameter modeling by conventional methods can be an intractable problem, involving deconvolution of instrument response, data model theory, model selection, and model refinement/fitting, which has prevented the realization of the technique's potential in the characterization of nano-dimensional thin film structures. This program addresses the mounting industry call for accuracy in thin film characterization (in particular; thickness, density, and roughness determination).

Technical Strategy

This fundamental XRR study involves two parallel characterization projects being performed on identical, temporally stable, multilayer artifacts supplied by SEMATECH. The NIST project consists of in-house XRR and HRXRD characterization with Système International (SI) traceable measurement instrumentation and SI traceable, first principles data modeling including Bayesian analysis providing refinement of instrumental and model parameters as well as structural model selection. In parallel to this NIST measurement effort, SEMATECH will measure or have measurements performed using commercial "in-line" XRR instrumentation and NIST will analyze these data using commercial software to address limitations to commercial instrumentation and software modeling. Combining results from both studies will ultimately allow accuracy traceability and error estimation for commercial instrumentation. This work will also address theoretical XRR modeling limitations and compare software model refinement and model selection approaches. The multi-year collaboration will provide the community with total error budget estimations for a given XRR structural analysis approach and instrumentation. This work will take approximately three years to address the issues discussed [FY's 05, 06, and 07]. The progress in FY05 includes a comprehensive, first principles development of the XRR theory necessary for traceable modeling of data and a first round of measurements on artifacts by commercial and NIST instrumentation.

NIST XRR Study – The NIST project consists of two parallel research components: 1) the development of SI traceable measurement instrumentation, 2) the creation of an SI traceable, first principles data analysis approach (see Fig. 1).

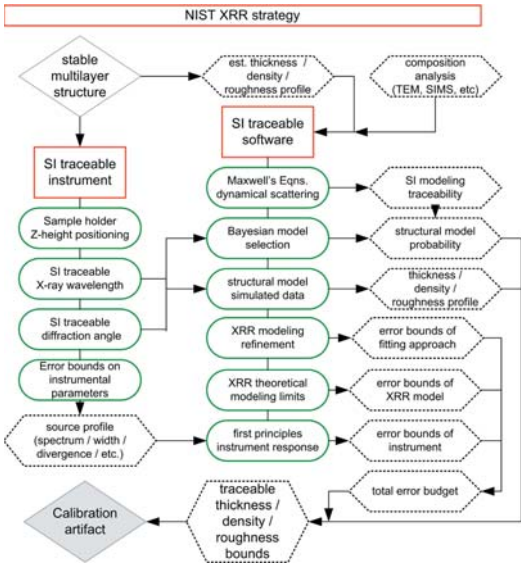

Figure 1. NIST XRR strategy. Measurements and modeling of temporally stable test structures will be performed on the NIST Ceramcis Divisiton Parallel Beam Diffractometer – an SI traceable instrumentation constructed for certification of XRD, HRXRD, and XRR Standard Reference Materials.

NIST has made previous efforts in developing in-house instrumentation capable of SI traceability for conventional XRD, HRXRD, and XRR techniques. Over the past seven years, NIST has constructed the Ceramics Division Parallel Beam Diffractometer (CDPBD) for SI traceable measurements of Powder XRD, Epitaxial HRXRD, and thin-film XRR artifacts for the Standard Reference Materials (SRM) program (Fig. 2). SI traceability in lattice parameter (XRD) or film thickness (XRR), d, requires simultaneous traceability in X-ray wavelength, λ, and diffraction angle, θ, following Bragg's law of diffraction: $2d = n\lambda / \sin(\theta)$.

Figure 2. Ceramics Division Parallel Beam Diffractometer (CDPBD). Showing the divergent X-ray source (center left), monochromator for generating parrallel/monochromatic X-rays (upper center), rotation stages for measuring diffraction angles (lower right) and receiving optics/detector (center right). In order to achieve accuracy in angle measurements and X-ray wavelength stability, numerous design features of varying complexity are present ranging from a "floating" platform holding the X-ray source to a hanging second rotation stage designed specifically to rotate the cables for the diffraction rotation stages.

The present NIST instrument development project involves addressing SI traceability aspects for both the diffraction angle and wavelength measurement in the CDPBD. Establishing SI traceability of the diffraction angle requires implementing optical encoding on the two rotation stages used to move the sample and detector. The optical encoder errors are then "mapped" using an external angle reference to generate SI traceability error bounds for each axis. The rotation stages presently have accuracy bounds of ± 2.0 μrads (± 0.4 arc seconds). Calibration experiments and collaboration with encoder manufacturers is currently underway to achieve an approximate one order of magnitude improvement in accuracy by next year [FY06 deliverable]. Determining SI traceability in wavelength involves constructing a stable, well modeled optics assembly to convert angular and energy divergent radiation from an X-ray source into parallel, monochromatic radiation for use in diffraction. The CDPBD uses a monochromator with Si (220), 2/4-bounce channel-cut crystals to filter the direct source into a source of highly parallel, single energy X-rays. SI traceability in X-ray wavelength from our source will be performed using an SI traceable reference crystal (with measured relative uncertainty of 3 in 10^{-8}). The X-ray wavelength for our instrumentation will have a relative uncertainty of ≈1 in 10^{-7} [FY06 deliverable]. The accuracy error bounds and instrument parameters from the wavelength and angle determination studies will provide an overall instrument response function which will be incorporated into NIST XRR profile modeling. The same instrumental parameters will establish guidelines for response profile modeling of commercial instrumentation [FY07 deliverable].

NIST is currently developing first principles, SI traceable XRR software to analyze data and refine model and instrument response parameters. XRR analysis is essentially an "inverse problem" wherein we select input parameters for a "guessed" structural model. This model is then used to simulate data that is compared with measured data and "Goodness of Fit" parameters are determined. This process is repeated until a "best fit" or best "Goodness of Fit" is found and the "best fit" model parameters become the "refined" model parameters used to describe the real structure. Three major questions limit the effectiveness of this current XRR modeling approach: 1) How do we accurately simulate the data using a structural model? 2) How do we know which structural model describes the

measured structure? 3) How do we accurately compare simulated and measured data? The NIST software development effort will attempt to answer each of these questions.

To address the model data simulation issue, we are currently developing XRR theory from Maxwell's equations and explaining all approximations and constraints required for the XRR modeling method. This approach will combine XRD, HRXRD and XRR in the same fundamental modeling theory and allow for SI traceability in derived refined parameters [FY05 deliverable]. To address the question of model accuracy, we are implementing a Bayesian/Maximum Entropy analysis approach to determine the probability that a given structural model correctly describes a given data set. This model selection component is essential to determining the validity of initial structural assumptions in any XRR analysis [FY06 deliverable]. Addressing the third question is of considerable interest with current commercial refinement approaches which often use dissimilar "chi squared" or model/simulation minimization criterion for overall refinement "figures of merit" such that comparison between refinement software packages is impossible. NIST will perform a rigorous analysis of current refinement approaches (such as genetic algorithms which are popular in commercial software for their speed advantage, Markov Chains Monte Carlo, simulated annealing, etc.) and of applicable minimization criterion to develop a consistent refinement approach for NIST modeling [FY07 deliverable].

NIST/SEMATECH XRR Study – The NIST/SEMATECH project consists of measurements and analysis from commercial instrumentation to allow comparison and calibration transfer from NIST SI traceable measurements and analysis (Fig. 3).

The NIST/SEMATECH project combines measurements with commercial "in line" XRR instrumentation with complimentary compositional analysis results as feedback for comparison with NIST measurements performed in parallel on the same artifacts. The measurement and modeling work at NIST will provide SI traceable XRR measurements and parameter analysis for artifacts and potential candidate SRMs. The collaboration work with SEMATECH allows interface and calibration transfer between NIST SI traceable measurements and "in-line" instrumentation. NIST will then use results from NIST SI traceable measurements, SEMATECH commercial

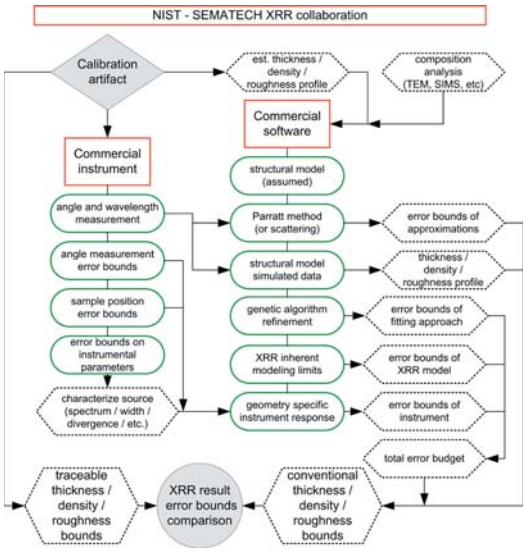

Figure 3. NIST – SEMATECH XRR strategy. Measurements of temporally stable test structures at SEMATECH will be performed on "fabrication-line-based" instrumentation and then analyzed at NIST using commercial software. These results will then be compared with NIST traceable results to provide calibration of commercial instrumentation.

measurements and modeling, and NIST SI traceable XRR modeling, to quantify error bounds and overall error budgets on the accuracy and precision possible from commercial instrumentation and commercial modeling software.

Calibration and accuracy determination of commercial instrumentation are the primary goals of this collaboration. To achieve these goals, we need to develop accurate instrument response functions for commercial instrumentation being calibrated. The instrument response function for commercial instrumentation may be the dominant term in the overall accuracy budget for XRR measurements. To address instrumentation effects on accuracy, work must be performed with different commercial XRR system geometries to discover the mechanical alignment parameters that dominate the error budgets of each system. This will require cooperation from instrument vendors to provide the necessary information or provide detailed instrument response functions directly. The incorporation of instrument response parameters into NIST modeling will allow for comparison between traceable measurements at NIST and measurements on commercial instrumentation in the field. The NIST instrument response function information will be developed through our instrument traceability study discussed earlier. We can then combine specific instrument "corrections"

to the accuracy error budget and use a calibration artifact such as a temporally stable thin film structure measured on the commercial instruments and at NIST to provide instrument calibration and stability monitoring [FY07+ deliverable].

Project Deliverables – The final project results available to SEMATECH will include XRR error budget estimations based on NIST XRR software and the calibration artifacts necessary for optimizing the performance of commercial "in-line" and laboratory XRR instruments. Accuracy and precision estimations provided by NIST software will determine the limitations of XRR applicability for arbitrary multilayer systems on commercial XRR instrumentation. Calibration artifacts measured on NIST SI traceable instrumentation will allow routine system monitoring, alignment calibration, and instrument response function comparisons to ensure commercial instrument precision and stability. These deliverables will dramatically improve the precision and accuracy of conventional XRR characterization of multilayer structures which exhibit well-established composition and uniformity [FY07+ deliverables].

Our deliverables schedule for the calendar year 6/2005-6/2006 time frame follows:

DELIVERABLES: First Principles roughness analysis. 1Q 2006

DELIVERABLES: Approximate Bayesian model selection method. 2Q 2006

DELIVERABLES: NIST analysis of SEMATECH measurements. 2Q 2006

DELIVERABLES: Report comparison of measurement results from ISMT and NIST instruments on same artifacts. 2Q 2006

Accomplishments

The CDPBD has recently been moved to our new equipment space in the Advanced Measurements Laboratory (AML at NIST) which is currently providing instrument temperature stability of ± 0.02 °C. Preliminary calibration of the angle encoders requires enhancements to current control electronics [4Q 05] to achieve target accuracy [by FY06]. First principles XRR theory development with emphasis on justifying approximations present in commercial XRR software will be completed this year [2Q 05]. Preliminary measurements of SEMATECH artifacts measured by both conventional and NIST traceable XRR systems will be completed this year [2Q 05].

FY05 Technical transfer: minimum thickness determination – Preliminary studies into theoretical limitations of XRR modeling have been completed this spring [2Q FY05] and have provided an approach to establishing the lower thickness limit for which the XRR technique provides useful results. First, data are simulated for a structure similar to the material that is being measured. Second, this simulated data are refined using commercial software. Third, the deviation between simulation input and model output indicates where XRR refinement of real data will fail. This approach can include actual measurement "noise" and real data collection ranges, so that the limits of different instrumentation effects and measurement times can also be analyzed. Figure 4 shows a study result for simulated HfO_2 on Si showing the regime where modeling fails to refine simulation thickness. Deviation provides the "minimum thickness" for which the XRR technique can be used for analysis in this structure. For the measured scan range, modeling fails for thickness < 0.8 nm.

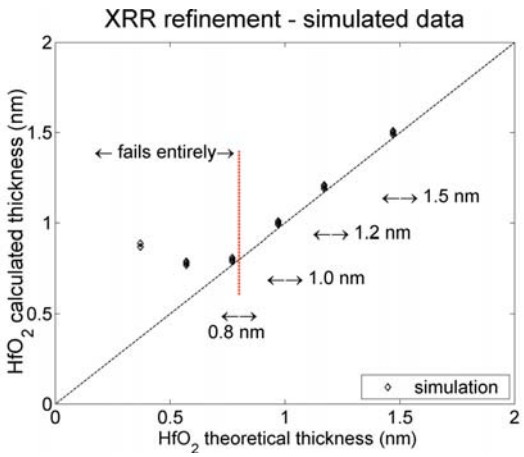

Figure 4. NIST Empirical determination of "minimum thickness." Simulated data which closely mimics a structure to be measured is modeled using commercial XRR refinement software. The thickness where modeling fails to determine the simulation thickness provides the "minimum thickness" where XRR analysis will provide accurate results on measured data. This analysis must be performed for each structure to be measured, and for the data range/noise levels present in the measured XRR data.

Collaborations

ISMT, Metrology – P.Y. Hung & Alain Diebold

PTB, Precision Metrology – Peter Thomsen-Schmidt

Bede Scientific Inc. – Keith Bowen & Matthew Wormington

Bruker AXS – Arnt Kern & Alan Coelho

Jordan Valley Semiconductors – Dileep Agnihotri

Technos International – Henry Yeung

Recent Publications and Presentations

D. Windover, N. Armstrong, J. P. Cline, P. Y. Hung, and A. Diebold, "Characterization of Atomic Layer Deposition using X-Ray Reflectometry" – poster/paper at 2005 International Conference on Characterization and Metrology for ULSI Technology, Dallas, TX, March, 2005.

N. Armstrong, W. Kalceff, J. P. Cline, and J. Bonevich, "A Bayesian/Maximum Entropy method for certification of a nanocrystallite-size NIST Standard Reference Material" – Ch. 8 in Diffraction analysis of the microstructure of materials, pp. 187-227, Ed. P. Scardi and E.J. Mittemeijer, Springer-Verlag, Berlin, ISBN: 3-540-40519-4, 2004.

N. Armstrong and P. Lynch, "Determining the dislocation contrast factor for X-ray line profile analysis" – Ch. 10 in Diffraction analysis of the microstructure of materials, pp. 249-307, Ed. P. Scardi and E.J. Mittemeijer, Springer-Verlag, Berlin, ISBN: 3-540-40519-4, (2004).

N. Armstrong, W. Kalceff, J. P. Cline, J. Bonevich, "Bayesian inference of nanoparticle-broadened X-ray line profiles" – J. NIST Research, 108(5), pg 155-178, (2004).

N. Armstrong, M. Leoni, M. Scardi, "Some concerns regarding the Wilkens' model for dislocation line broadening" – invited presentation at Size-Strain 4: EPDIC 9, Prague, Czech Republic, September, 2004.

N. Armstrong, J. P. Cline, W. Kalceff, and A. Dowd, "Status of the NIST Nanocrystalline Size SRM - 1979" – invited presentation at Size-Strain 4: EPDIC 9, Prague, Czech Republic, September, 2004.

N. Armstrong, J. P. Cline, W. Kalceff, and A. Dowd, "Bayesian characterization of nanoparticles from line profile" – presentation at Denver X-Ray Conference, Denver, CO, August, 2004.

N. Armstrong, W. Kalceff, J. P. Cline, J. Bonevich, P. A. Lynch, C. C. Tang, S. A. Thompson, "X-ray diffraction characterization of nanoparticle size and shape distributions: -- Application to bimodal distributions" – Proceedings of the 28th Annual Condensed Matter & Material Meeting, Wagga, Australia, February, 2004.

J. P. Cline and S. T. Misture, "Alignment & Standards" – workshop at Denver X-Ray Conference, Denver, CO, August, 2003.

D. Windover and J. P. Cline "Goniometer angle calibration for traceability using the method of circle closure" – poster at Denver X-Ray Conference, Denver, CO, August, 2003.

D. Windover and J. P. Cline, "Calibration standards for X-ray metrology systems using a traceable high-resolution diffractometer" – poster at American Vacuum Society, Baltimore, MD, November, 2003.

Electron Microscope Tomography of Electronic Materials

Technical Contacts:
Z. H. Levine
J. H. Scott

Goals

Enable the use of three-dimensional imaging for thick samples using commercial scanning transmission electron microscopes (STEM). Typical samples include porous low-κ dielectrics, two-layer interconnect samples, and photonic band gap materials.

Customer Needs

The NTRS/ITRS has recognized the need for three-dimensional imaging of interconnects for several years. In this study, our principle objective is to determine the morphology of pores in low-κ dielectric material. Two aspects of the pore distribution are critical: (a) the largest pores may lead to failure of the dielectric (*e.g.*, short circuits), and (b) the connectivity of the pores is important to understand the transport of chemicals during the fabrication of the interconnect.

The potential solutions and major challenges for interconnect are discussed in the 2004 International Technology Roadmap for Semiconductors Update on pages 2 and 17 of the Interconnect Section. Minimization of size effects, 3-D characterization of low-κ void intersection with sidewalls, pore size distribution and barrier roughness are some of the important issues in porous low-κ measurements.

Technical Strategy

1. First, we will upgrade an existing commercial transmission electron microscope to be able to obtain high angle STEM images with a full quantitative understanding of the input and output signals. Noise reduction is also a key issue.

DELIVERABLES: Hardware fabrication for extraction of bright field and dark field STEM detector amplifier chain gain and offset. 2Q 2005

2. In parallel, we will develop theoretical and computational expertise for the understanding of STEM signals associated with multiple scattering of electrons.

DELIVERABLES: 3-D Monte Carlo simulation code in Java and Jython for prediction of energy-dispersive X-ray signals with full 3-D geometrical complexity including electron transport and X-ray absorption effects. 1Q 2005

3. We will obtain a tilt series and perform a tomographic — analysis of a photonic band gap system an artificially periodic polymer structure.

4. We will perform a similar analysis on a low-κ dielectric material.

DELIVERABLES: Code for 3-D Reconstruction with Bayesian Statistics for the Projective Multiple Scattering Regime. 2Q 2005.

Levine (2003) demonstrated by computer simulation that it was possible to reconstruct polymer samples with a thickness well in excess of 1 micrometer if multiple scattering effects were accounted for. In this project, we will generalize the Bayesian tomographic reconstruction algorithm known as the Generalized Gaussian Markov Random Field, introduced by Bouman and Sauer in 1993, to generalize the transmission function from Beer's law to a general function of the integral of a material parameter, i.e., to permit the inclusion of multiple scattering effects. Also, the program will run in 3-D, rather than 2-D in the Bouman-Sauer implementation.

DELIVERABLES: Tomographic study of a low-κ dielectric material. 3Q 2005

The porous low-κ dielectric will pose more of a challenge, particularly for alignment, because of its random nature. Hence, it will be studied second.

Accomplishments

- To date, we demonstrated the ability to obtain three-dimensional information using a scanning confocal transmission electron microscope on integrated interconnect samples several micrometers (Frigo, Levine, and Zaluzec, 2002) as well as the ability to obtain three-dimensional information on integrated circuit interconnect samples using a commercial scanning transmission electron microscope (Levine *et al.*, 2003).

- We performed a 3-D reconstruction of a photonic crystal supplied by Lucent Technologies (see Fig. 1, pg. 152), based on data taken on our in-house STEM.

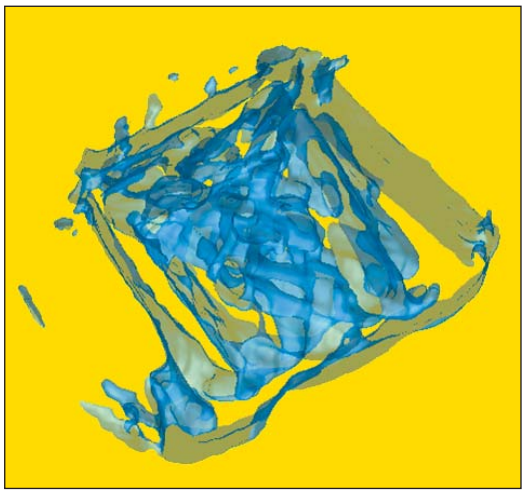

Figure 1. Tomographic reconstruction of a photonic band gap sample. An interconnecting network of material and void is present in the center of the figure. The bands on the outside are from the platinum overlayer added to protect the sample during preparation by a focused ion beam.

■ We simulated the observed STEM scattering and energy-dispersive X-ray signal from a 3-D phantom containing interconnect materials over an angular range of 180 degrees (Fig. 2).

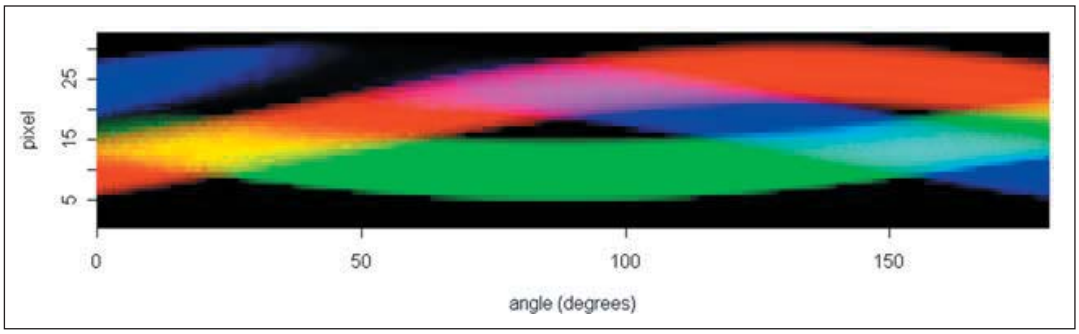

Figure 2. 3-D Java Monte Carlo simulation of interconnect phantoms and observed X-ray emission (Cu=red Al=blue SiO2=green) from 0 degree to 180 degree tilt.

Collaborations

International Sematech, Brendan Foran, preparation of low-κ samples, electron microscopy.

Chris Soles and Hae-Jeong Lee, NIST, MSEL, Polymers Division; low-κ samples

Accurel, Inc. Preparation of focused ion beam sections from low-κ samples

Lucent Technologies, Shu Yang; preparation of photonic band gap material.

Recent Publications

S, Frigo, Z. H. Levine, and N. J. Zaluzec, "Submicron imaging of buried integrated circuit structures using a scanning confocal electron microscope," Appl. Phys. Lett. **81**, 2112 (2002).

Z. H. Levine, "Tomography in the Multiple Scattering Regime of the Scanning Transmission Electron Microscope," Appl. Phys. Lett. **82**, 3943 (2003).

Z. H. Levine, J. A. Gao, S. Neogi, T. M. Levin, J. H. Scott, and S. Grantham, "Parallax measurements of an integrated circuit interconnect using a scanning transmission electron microscope," J. Appl. Phys. **93**, 2193 (2003).

Z. H. Levine, S. Grantham, D. J. Patterson, I. McNulty, I. C. Noyan, and T. M. Levin, "Imaging material components of an integrated circuit interconnect," J. Appl. Phys. **95,** 405 (2004).

Z. H. Levine, "An X-Ray Tomography Primer," Electronic Device Failure Analysis **7**, 26 (2005).

Z. H. Levin e, "Theory of Bright-Field Scanning Transmission Electron Microscopy," J. Appl. Phys. **97**, 033101 (2005).

High-Resolution Microcalorimeter X-Ray Spectrometer for Chemical Analysis

Technical Contacts:
J. N. Ullom
J. A. Beall
W. B. Doriese
W. D. Duncan
G. C. Hilton
K. D. Irwin
S. W. Nam
C. D. Reintsema
L. R. Vale

Goals

We will develop new generations of X-ray spectroscopy tools to meet the materials analysis needs of the semiconductor manufacturing industry. Energy-Dispersive Spectrometers (EDS) based on microcalorimeters have the ability to detect photons with high energy resolution and near-unity quantum efficiency. Using these tools, a wide range of materials analysis problems can be solved. In semiconductor manufacturing, improved X-ray materials analysis is needed to identify nanoscale contaminant particles on wafers and to analyze very thin layers of materials and minor constituents. Microcalorimeter EDS improves the spectral resolution by one to two orders of magnitude compared to the semiconductor Si-EDS, the existing industry standard. Such improved resolution combined with energy dispersive operation makes possible direct spectral separation of most overlapping peaks often encountered with Si-EDS in complex multi-element systems. The improved resolution of the microcalorimeter EDS also increases the peak-to-background ratio. Peak shape and shift can be studied to reveal chemical state information.

Developing arrays of X-ray microcalorimeters will enable the acquisition of high statistics spectra in reduced time, improving the efficiency and statistical quality of existing materials analysis applications. Further, large-format arrays (up to 1000 pixels) will make it possible to chemically analyze smaller features and trace constituents, and to track rapidly evolving X-ray spectra for in-process and process-stream monitoring.

The microcalorimeter EDS detector invented at NIST consists of a superconducting thermometer (a superconducting transition-edge sensor (TES)) and an X-ray absorber fabricated on a micromachined Si_3N_4 membrane, and cooled to cryogenic temperatures (0.1 K). When X-rays are absorbed in the detector, the resulting heat pulse in the microcalorimeter is measured by the TES thermometer. The change in the temperature of the thermometer is measured by a superconducting quantum interference device (SQUID) amplifier. The temperature pulse height gives a measurement of the energy of the X-ray photon one to two orders of magnitude more sensitive than Si-EDS. The detector is cooled to 0.1 K by a compact adiabatic demagnetization refrigerator.

Customer Needs

Improved X-ray detector technology has been cited by SEMATECH's Analytical Laboratory Managers Working Group (ALMWG, now Analytical Laboratory Managers Council (ALMC) as one of the most important metrology needs for the semiconductor industry. In the International Technology Roadmap for Semiconductors, improved X-ray detector technology is listed as a key capability that addresses analysis requirements for small particles and defects. The transition-edge sensor (TES) microcalorimeter X-ray detector developed at NIST has been identified as a primary means of realizing these detector advances, which will greatly improve in-line and off-line metrology tools that currently use semiconductor energy-dispersive spectrometers (EDS). At present, these metrology tools fail to provide fast and unambiguous analysis for particles less than approximately 0.1 μm to 0.3 μm in diameter. Improved EDS detectors such as the TES microcalorimeter are necessary to extend the capabilities of existing SEM-based instruments to meet the analytical requirements for future technology generations. With continued development, microcalorimeter EDS should be able to meet both the near-term and the longer-term requirements of the semiconductor industry for improved particle analysis.

"Promising new technology such as high-energy resolution X-ray detectors must be rapidly commercialized. Prototype microcalorimeter energy dispersive spectrometers (EDS) and superconducting tunnel junction techniques have X-ray energy resolution capable of separating overlapping peaks and providing chemical information. These advances over traditional EDS and some wavelength dispersive spectrometers can enable particle and defect analysis on SEMS located in the clean room." **2003 International Technology Roadmap for Semiconductors**

Technical Strategy

1. The usefulness of single-pixel X-ray microcalorimeter EDS in materials analysis has now been well established in a variety of demonstrations. Arrays of X-ray microcalorimeters are being tested, and the development of technologies for large-format arrays is in process. To meet the needs of the semiconductor industry, it is nec-

essary to make both single-pixel and array microcalorimeter systems more widely available. In addition to microcalorimeters, the system requires novel superconducting electronics to instrument the detectors, compact adiabatic demagnetization refrigerators to simplify cooling to milliKelvin operating temperatures, and custom room-temperature electronics and software to process the output signals. Our goal is to develop new generations of detector systems, to transfer them to the Chemical Science and Technology Laboratory (CSTL) in NIST Gaithersburg for collaborative use in studying problems of interest to the industry (Fig. 1), and to work with other partners in disseminating the technology.

Figure 1. Single-pixel NIST X-ray microcalorimeter system on a scanning-electron microscope. The system was developed in the Electronics and Electrical Engineering Laboratory (EEEL) at NIST, Boulder, and transferred to the Chemical Science and Technology Laboratory (CSTL) at NIST Gaithersburg to be used to study problems of interest to the semiconductor industry.

DELIVERABLES: Provide continued support and upgrades for the single-pixel microcalorimeter system transferred to CSTL. Prepare for the eventual transfer of an array microcalorimeter system. Collaborate with CSTL on using the microcalorimeter to study problems of interest to the semiconductor industry.

2. Continued improvements in microcalorimeter energy resolution are desirable for the rapid analysis of chemical shifts. However, for many semiconductor materials analysis problems, further improvements in energy resolution (beyond that already demonstrated with these detectors) are not as important as an increase in the maximum count rate and collection area. Both the collection area and count rate can be improved by the implementation of multipixel arrays of detectors. Arrays of X-ray microcalorimeters are now being fabricated and tested. The performance of these arrays should be similar to that of earlier, successful single pixels.

DELIVERABLES: Achieve high-resolution operation of a close-packed X-ray microcalorimeter array to demonstrate the increase in collection area and count rate achievable with arrays.

3. One of the barriers to widespread dissemination of X-ray microcalorimeter instruments is the complexity and cost of the adiabatic demagnetization refrigerator used to cool to 100 mK. The development of an on-chip solid-state microrefrigerator based on superconducting tunnel junctions to cool from 300 mK to 100 mK would greatly simplify the cryogenic system needed for microcalorimeter EDS. Adiabatic demagnetization refrigerators could be replaced by small, simple, and inexpensive ^{3}He systems, which cool to 300 mK, coupled to the solid-state tunnel-junction refrigerator cooling to 100 mK.

DELIVERABLES: Demonstrate the cooling of an electrically-separate piece of thin-film electronics using a tunnel-junction refrigerator that could be coupled to a simple ^{3}He refrigerator.

4. The operation of microcalorimeter arrays requires multiplexed SQUID readout. A time-domain SQUID multiplexer has previously been demonstrated. The integration of this multiplexer with X-ray microcalorimeters is a crucial step towards the deployment of large sensor arrays. The operation of such an integrated system under X-ray illumination will allow its capabilities to be measured and routes to improvement to be determined.

DELIVERABLES: Demonstrate multiplexed operation of microcalorimeters under X-ray illumination. Characterize multiplexer performance and design next-generation system.

Accomplishments

- We fabricated and tested a new generation of microcalorimeters that incorporate additional normal metal regions to suppress internal noise. In addition to being more stable and easier to bias, the energy resolution of these microcalorimeters is significantly improved. We demonstrated a world record energy resolution of 2.4 eV FWHM at 5.9 keV (see Fig. 2).

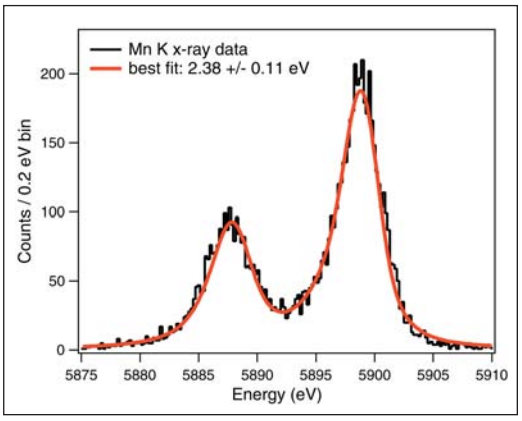

Figure 2. X-ray spectrum from improved NIST microcalorimeter. The FWHM resolution was 2.4 eV for 5.9 keV X-rays.

■ We continue to support the microcalorimeter system installed on a CSTL scanning electron microscope in Gaithersburg, Maryland. We recently upgraded the infrared blocking filters, the magnetic shielding, the pulse acquisition hardware, the detector bias hardware, and the microcalorimeter itself. The new detector is more stable, easier to bias, and has better spectral resolution than the previous unit. We are collaborating with CSTL researchers on further improvements to the stability and ease-of-use of the system.

■ We recently demonstrated the ability to read out eight TES microcalorimeters in a single multiplexed amplifier channel with 3.8 eV FWHM energy resolution (Fig. 3). The detectors were fast, having a signal decay-time constant of about 150 microseconds. Combining this result with closed-form calculations and a new Monte-Carle software package that performs a detailed simulation of our SQUID multiplexer, we can reliability predict the future performance of the multiplexer system. In particular, with only evolutionary improvements to the basic architecture, our time-division SQUID multiplexer will be able to readout 32 detectors per channel with 4 eV resolution or better. Planned improvements include increased open-loop system bandwidth, well-matched pulse rise and fall times, lower SQUID noise, and optimization of the coupling between microcalorimeters and SQUIDs. We are now preparing to demonstrate multiplexed operation of a 3x32 high-resolution microcalorimeter array.

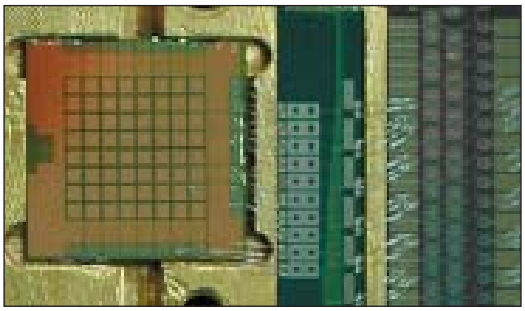

Figure 3. Photograph of an 8x8 array of microcalorimeters. Each microcalorimeter has a 1.5-μm-thick Bi absorber. The middle chip is a filter chip. The chip on the right is a SQUID multiplexer.

■ We have continued work on an on-chip solid-state refrigerator to cool X-ray microcalorimeters from 300 mK to 100 mK. If successful, this refrigerator could greatly simplify the cryogenic system needed for microcalorimeter EDS. Adiabatic demagnetization refrigerators could be replaced by small and inexpensive ^{3}He systems coupled to the solid-state refrigerator. The device is a superconducting analog of a Peltier cooler. Cooling is produced in the devices by the tunneling of electrons through normal-insulator-superconductor junctions. We have previously demonstrated refrigerators able to cool themselves from 260 mK to 130 mK with cooling powers well matched to microcalorimeter X-ray sensors. Recently, we demonstrated refrigerators able to cool bulk material as well as electrically separate pieces of thin-film electronics, such as a X-ray microcalorimeter (Fig. 4). The solid-state refrigerator is

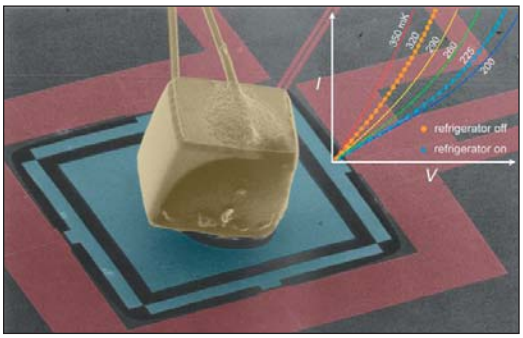

Figure 4. Colorized micrograph of solid-state refrigerator. A cube of germanium (yellow) located on a suspended, micromachined membrane is cooled by four pairs of normal-insulator-superconductor tunnel junctions. The ratio of the volumes of the germanium and the junctions is the same as the ratio of the volumes of the Statue of Liberty and an ordinary person (about 11,000). Macroscopic wires (at upper left) contact the germanium to measure its resistance; these are cooled as well.

fabricated on a silicon wafer using conventional thin-film and photolithographic techniques, has no moving parts, and operates continuously. We recently cooled a thin-film payload from 320 mK to 225 mK. This work has been featured twice on the cover of *Applied Physics Letters* and once on the cover of *Physics Today*.

■ The NIST microcalorimeter EDS holds the world record for energy resolution for an EDS X-ray detector of 2.0 eV at 1500 eV, which is over 30 times better than the best high resolution semiconductor-based detectors currently available (Fig. 5). This energy resolution was measured using a glass prepared by Dale Newbury of NIST to use as a test standard for EDS.

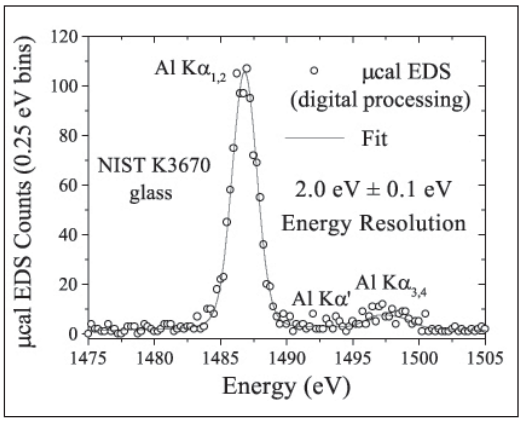

Figure 5. The Al-Kα region of a microcalorimeter EDS spectrum of the multielement NIST K3670 glass. The acquired spectrum is shown as well as a weighted least-squares fit of the Al Kα and satellite lines convolved with a Gaussian instrument response, yielding an energy resolution of 2.0 eV ± 0.1 eV FWHM.

■ We created a chemical shift map showing the chemical bonding state of Al in a sample containing both Al and Al_2O_3 (see Fig. 6). An aluminum film was deposited on part of a sapphire substrate. A microcalorimeter EDS was used to measure the X-ray spectrum as the electron beam was rastered to form the SEM image. The Al X-ray line position was shifted by a small amount (about 0.2 eV) in the regions containing Al_2O_3 as compared to the regions containing elemental Al. The high energy resolution of the microcalorimeter allowed the shift to be measured, resulting in the false-color image below. The map clearly demonstrates that microcalorimeter EDS can be used to discriminate the chemical bonding state using shifts in the positions of X-ray lines. The result also highlights the need for large-format arrays to increase the data-collection rate. The image shown here was acquired over several hours. Images such as this could be acquired much more quickly and with much sharper position resolution using an array microcalorimeter.

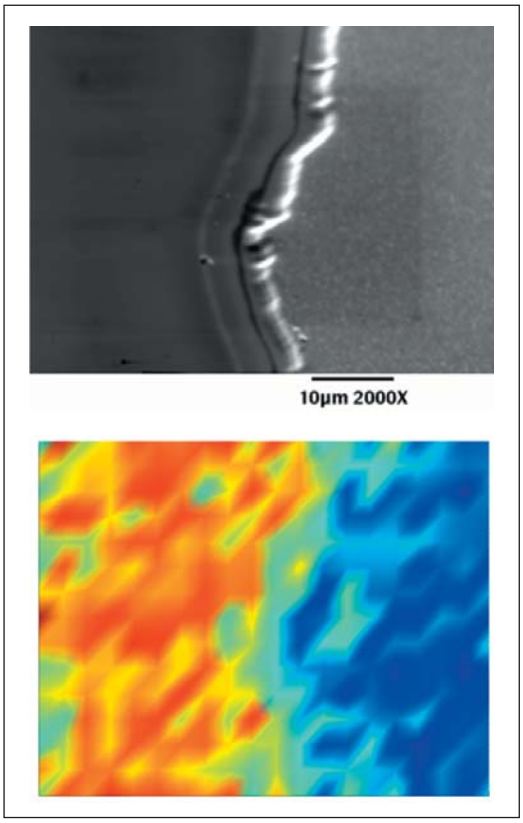

Figure 6. SEM photo (top) of an aluminum film partially covering a sapphire substrate and a false-color map (bottom) of the shift in the mean X-ray energy of the Al Kα peak taken using a microcalorimeter EDS system. The shift in energy is due to the chemical bonding of the aluminum with the oxygen. The average energy shift is ~0.2 eV.

Collaborations

Chemical Science and Technology Laboratory, NIST, Gaithersburg, Terry Jach, Lance King, Dale Newbury, John Small, and Eric Steel, the development of microcalorimeter EDS systems.

Recent Publications

J. N. Ullom, J. A. Beall, W. B. Doriese, W. D. Duncan, L. Ferreira, G. C. Hilton, K. D. Irwin, C. D. Reintsema, L. R. Vale, "Optimized Transition-Edge X-ray Microcalorimeter with 2.4 eV Energy Resolution at 5.9 keV," Submitted to Appl. Phys. Lett.

A. M. Clark, N. A. Miller, A. Williams, S. T. Ruggiero, G. C. Hilton, L. R. Vale, J. A. Beall, K. D. Irwin, J. N. Ullom, "Cooling of Bulk Material by Electron-Tunneling Refrigerators," Appl. Phys. Lett. 86, 173508, (2005).

W.B. Doriese, J. A. Beall, S. Deiker, W. D. Duncan, L. Ferreira, G. C. Hilton, K. D. Irwin, C. D. Reintsema, J. N. Ullom, L. R. Vale, and Y. Xu, "Time-division multiplexing of high-resolution TES X-ray microcalorimeters: Four pixels and beyond," Appl. Phys. Lett. 86, 4762 (2004).

S. W. Deiker, W. B. Doriese, G. C. Hilton, K. D. Irwin, W. H. Rippard, J. N. Ullom, L. R. Vale, S. T. Ruggiero, A. Williams, and B. A. Young, "Transition edge sensor using dilute AlMn alloys," Appl. Phys. Lett., 85, 2137 (2004).

K. D. Irwin, K. W. Lehnert, "Microwave SQUID Multiplexer," Appl. Phys. Lett. 85, 2107 (2004).

J. Pekola, R. Schoelkopf, J.N. Ullom, "Cryogenics on a Chip," Physics Today, May 2004, 41.

J.N. Ullom, W. B. Doriese, G. C. Hilton, J. A. Beall, S. Deiker, W. D. Duncan, L. Ferreira, K. D. Irwin, C. D. Reintsema, L. R. Vale, "Characterization and Reduction of Unexplained Noise in Superconducting Transition-Edge Sensors," Appl. Phys. Lett., 84, 4206 (2004).

A.M. Clark, A. Williams, S. Ruggiero, M. L. van den Berg, J.N. Ullom, "Practical Electron-Tunneling Refrigerator," Appl. Phys. Lett. 84, 625 (2004).

S. Ruggiero, A. Williams, W. H. Rippard, A. Clark, S. Deiker, L. R. Vale, J.N. Ullom, "Dilute Al-Mn Alloys for Superconductor Device Applications," J. Low Temp. Phys. 134, 973 (2004).

S. Ruggiero, A. Williams, W. H. Rippard, A. Clark, S. Deiker, L. R. Vale, J.N. Ullom, "Dilute Al-Mn Alloys for Superconductor Device Applications," Nucl. Instrum. & Methods in Phys. Res. A 520, 274 (2004).

K. D. Irwin, D. Audley, J. A. Beall, J. Beyer, S. Deiker, W. B. Doriese, W. Duncan, G. C. Hilton, W. Holland, J.N. Ullom, L. R. Vale, Y. Xu, "In-focal-plane SQUID multiplexer," Nucl. Instrum. & Methods in Phys. Res. A 520, 544 (2004).

G. C. Hilton, J. A. Beall, S. Deiker, L. R. Vale, W. B. Doriese, J. Beyer, J.N. Ullom, C. D. Reintsema, Y. Xu, K. D. Irwin, "X-ray Microcalorimeter Arrays Fabricated by Surface Micromachining," Nucl. Instrum. & Methods in Phys. Res. A 520, 435 (2004).

W. B. Doriese, J. Beall, J. Beyer, S. Deiker, S. Ferreira, G. C. Hilton, K. D. Irwin, J. M. Martinis, S.W. Nam, C. D. Reintsema, J.N. Ullom, L. R. Vale, "A Time-Domain SQUID multiplexer for readout of X-ray microcalorimeter arrays," Nucl. Instrum. & Methods in Phys. Res. A 520, 559 (2004).

J.N. Ullom, W. B. Doriese, G. C. Hilton, J. A. Beall, S. Deiker, K. D. Irwin, C. D. Reintsema, L. R. Vale, Y. Xu, "Suppression of excess noise in transition-edge sensors using magnetic field and geometry," Nucl. Instrum. & Methods in Phys. Res. A 520, 333 (2004).

N. A. Miller, J.N. Ullom, J. A. Beall, G. C. Hilton, S. Deiker, W. B. Doriese, K. D. Irwin, C. D. Reintsema, L. R. Vale, Y. Xu, "Robust infrared filters for X-ray spectroscopy," Nucl. Instrum. & Methods in Phys. Res. A 520, 638-640 (2004).

B. L. Zink, K. D. Irwin, G. C. Hilton, D. P. Pappas, J.N. Ullom, M. Huber, "Lithographically patterned magnetic calorimeter X-ray detectors with integrated SQUID readout," Nucl. Instrum. & Methods in Phys. Res. A 520, 52 (2004).

C. D. Reintsema, J. A. Beall, S. Deiker, W. B. Doriese, G. C. Hilton, K. D. Irwin, S.W. Nam, L. R. Vale, J.N. Ullom, Y. Xu, J. Beyer, M. Huber, "A Time-Domain SQUID Multiplexer System for Read-Out of Superconducting Transition-Edge-Sensor Arrays," Ext. Abstracts, 2003 Intl. Superconductive Electronics Conf. (ISEC), Jul 07-12, 2003, Sydney, Australia.

C. D. Reintsema, J. Beyer, S.W. Nam, S. Deiker, G. C. Hilton, K. D. Irwin, J. M. Martinis, J.N. Ullom, L. R. Vale, M. MacIntosh, "A Prototype System for Time-Domain SQUID Multiplexing of Large-Format Transition-Edge Sensor Arrays," Rev. Sci. Instrum 74, 4500 (2003).

Piet A. J. de Korte, Joern Beyer, Steve Deiker, Gene C. Hilton, Kent D. Irwin, Mike MacIntosh, Sae Woo Nam, Carl D. Reintsema, Leila R. Vale, and Martin E. Huber, "Time-division superconducting quantum interference device multiplexer for transition-edge sensors," Rev. Sci. Instrum. 74, 3807 (2003) .

K. D. Irwin, J. A. Beall, J. Beyer, S. Deiker, W. B. Doriese, S. Ferreira, G. C. Hilton, S.W. Nam, C. D. Reintsema, J.N. Ullom, L. R. Vale, "Superconducting Multiplexers for Transition-Edge Sensors," International Progress on Advanced Optics and Sensors, Universal Academy Press, Inc., pp. 437-442.

K. D. Irwin, J. A. Beall, J. Beyer, S. Deiker, W. B. Doriese, S. Ferreira, G. C. Hilton, S.W. Nam, C. D. Reintsema, J.N. Ullom, L. R. Vale, "Time-Division SQUID Multiplexers for Transition-Edge Sensors," Proc. Intl Wkshp on Extreme Optics and Sensors, Jan 14-17, 2003, Tokyo, Japan, pp. 195-196.